Exotic fission properties of highly neutron-rich Uranium isotopes

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The series of Uranium isotopes with $N = 154 \sim 172$ around the magic number N=162/164 are identified to be thermally fissile. The thermal neutron fission of a typical representative ^{249}U of this region amenable to synthesis in the radioactive ion beam facilities is considered here. Semiempirical study of fission barrier height and width shows this nucleus to be infinitely stable against spontaneous fission due to increase in barrier width arising out of excess neutrons. Calculation of probability of fragment mass yields and microscopic study in relativistic mean field theory, show this nucleus to undergo a new mode of thermal fission decay termed *multifragmentation fission* where a number of prompt scission neutrons are simultaneously released along with the two heavy fission fragments.

Exploration of nuclear landscape with radioactive ion beam facilities is currently underway in several laboratories around the world. Presently about 2000 nuclei are known, and different mass models predict the survival of another 5000 nuclei with varying life time which could be synthesised in the laboratory [1]. Most of the nuclei will occur in the neutron-rich side of the nuclear chart. Many of these nuclei will have unusual neutron to proton ratio, quite different from the usual ones in the valley of stability. Therefore with the synthesis of such nuclei one may expect to observe new nuclear phenomena and novel features of nuclear dynamics. The two Uranium isotopes ^{233}U and ^{235}U and Plutonium isotope ^{239}Pu in the actinide region are suitable for energy production being thermally fissile in nature. Here we investigate to see if heavier neutron-rich isotopes of Uranium could exist having thermally fissile properties, and if so what would be their fission decay properties.

On the basis of fission barrier B_f and neutron separation energy S_n systematics, we find that the chain of Uranium isotopes with neutron number N=154 ∼172 possess the thermally fissile property [2]. These isotopes span on either side of N=162/164 which has been predicted to be magic in numerous theoretical studies carried over the years [3]. The effect of close shell manifests in relatively higher values of S_n and lower values of B_f (favouring more bound compact configuration) rendering the system thermally fissile, which may be the case with the above series of Uranium isotopes. All these nuclei are stable against alpha decay and several of them in the lower side have beta-decay half-life of several tens of seconds [4]. We have chosen ^{249}U, which leads to ^{250}U with capture of thermal neutron for the present study. This isotope is only 11 neutrons away from the naturally occuring known Uranium isotope ^{238}U, and therefore likely to be produced with RIB facilities under construction. The question of how the neutron-rich isotopes of the actinide nuclei will decay by fission has not been addressed before. The neutron separation energy S_n provides a measure of excitation of the compound nucleus. In our study we use the fission barrier from the extensive study of Howard and Möller (HM) [2] which is being widely

TABLE I: Fission barrier B_f (in MeV) obtained in finite-range droplet model (FRDM) [5] and Howard and Möller (H.M.) [2] are compared with experimental data [5].

nuclei	B_f (expt.)	B_f (H.M.)	B_f (FRDM)
^{228}Th	6.50		7.43
^{230}Th	7.00	7.79	7.57
^{232}Th	6.30	7.40	7.63
^{234}Th	6.65	6.83	7.44
^{232}U	5.40	6.31	6.61
^{234}U	5.80	6.03	6.79
^{236}U	5.75	5.74	6.65
^{238}U	5.90	5.83	4.89
^{240}U	5.80	5.92	5.59
^{238}Pu	5.30	5.25	4.85
^{240}Pu	5.50	5.92	4.74
^{242}Pu	5.50	5.48	5.25
^{244}Pu	5.30	5.31	5.78
^{246}Pu	5.30	5.06	6.27
^{242}Cm	5.00	5.56	4.24
^{244}Cm	5.00	5.56	5.05
^{246}Cm	4.70	5.40	5.69
^{248}Cm	5.00	5.08	6.07
^{250}Cm	4.40	4.53	5.51

used in literature. It is also the only exhaustive calculation which gives barrier for the whole range of known and unknown nuclei where fission decay can occur. We have compared in Table I the HM values of B_f with experiment for 18 actinide nuclei of the four elements Th, U, Pu and Cm for which data are available [5]. These data are for the external barriers which in general correspond to the maximum barrier height calculated by Howard and Möller. We have also included in the table the B_f values calculated by Möller et al [5] in finite-range droplet model (FRDM) for a relative comparison. It is interesting to see that HM values are in much better agreement with experiment than those of FRDM, although the latter are comparatively more recent. For the Uranium isotopes which is of special interest in the present study, the agreement is remarkably satisfactory. Therefore we have used them in Fig. 1, shown as closed circles for all the

Uranium isotopes with the neutron number in the range $140 \sim 180$. For the neutron separation energies S_n, we have used the three mass formulae HM [2], FRDM [4] and infinite nuclear matter (INM) [6]. It is well known that if $S_n < B_f$, then the nucleus cannot undergo thermal neutron fission. The fission threshold $E_{nm} = B_f - S_n$ has to be overcome by impinging with an energetic neutron, in order that the nucleus will undergo fission. However, if $S_n > B_f$, then thermal neutron (with practically zero energy) can cause fission. It is interesting to see in Fig. 1 that, the isotopes of Uranium in the range $N = 154 - 172$ are thermally fissile, a feature emanating from the close shell nature of N=162/164. To show that this feature is not accidental or specific to Uranium, but a general one arising out of the close shell structure of N=162 (or 164), we have presented the case of Th-isotopes in the same figure.

We now consider the nature of the fission decay mode of ^{250}U, which is primarily governed by the profile of the fission barrier. The height and width of the fission barrier which is supposed to be parabolic in nature have to be obtained[7]. We have followed a semiempirical method to get the width of the barrier from the systematics of the known experimental fission half-lives, and extrapolated them to neutron-rich region of interest. In spontaneous fission the fissioning fragment will see the maximum barrier which will determine the tunneling probability more decisively. Hence, the maximum barrier given by HM [2] can be considered as the effective parabolic barrier governing the decay, which should be appropriate for the present study. The fission half-lives can be calculated using the relation [7] $\tau_{1/2} = ln2/np$, where n is the number of barrier assaults by the decaying fragment related to the barrier curvature energy $\hbar\omega$ by $n\hbar = \hbar\omega/2\pi$, and p is the penetrability of the barrier given by $p = [1 + exp(2\pi B_f/\hbar\omega]^{-1}$. It may be noted that $\hbar\omega$ is a measure of width of the barrier; smaller the $\hbar\omega$ larger is the barrier width, and hereafter we refer it as width to bring out a physical picture. Taking the HM values [2] of B_f and using the experimental $\tau_{1/2}$ [8], we get the values of $\hbar\omega$. The systematics of $\hbar\omega$ versus B_f so obtained are shown in Figure 2 for various even-even actinide nuclei. It is indeed quite revealing that the plot shows a linear behaviour of $\hbar\omega$ with B_f for a given Z, with progressively increasing slope with the increase of proton number of the elements from 90 to 96. For the next element Cf with Z=98, the linear behavior gets fuzzy. However, the mean follows the trend with a higher inclination. The trend is more conspicuously restored for Fm with Z=100. This deviation coincidentally correlates well with the fission mass yield systematics, where considerable deviation from the standard well-defined two peaks occurs for Fm isotopes. The width $\hbar\omega$ for any isotope with calculated fission barrier may be obtained by extrapolation of the linear graph. Since ^{250}U has 158 neutrons and of considerable distance from the close shell N=162 (or 164) and itself only 12 neutrons away from the known ^{238}U this extrapolation should be quite meaning-

ful and reliable. For ^{250}U with a fission barrier of 4.3 MeV [2], we obtained the value of $\hbar\omega$ as 0.225 MeV from Fig. 2. Now we can construct the parabolic barrier of base width $\triangle r$ somewhat schematically, using the value of $\hbar\omega$ following the relation $1/\hbar\omega = d^2V/dr^2$, where V is the potential energy. The barrier so obtained is relatively flat and wide compared to the fission barrier of ^{236}U with $\hbar\omega = 0.357$ MeV, obtained from the experimental half-life [8] and $B_f = 5.74$ MeV from Howard and Möller [2]. The normal nuclei like ^{230,232}Th or ^{235,238}U have $\hbar\omega \approx 0.4 - 0.5$MeV. Thus, the neutron-rich heavy isotope ^{250}U has considerably lower value of $\hbar\omega$, and consequently relatively larger width, arising out of excess of neutrons. This flattening of fission barrier makes the nucleus stable against spontaneous fission decay, because of decreasing penetrability. The spontaneous fission decay half-life calculated with these height and width comes out to be 5.7×10^{24} years, which is several order of magnitude higher than that of ^{236}U. However, due to the decreased fission barrier of 4.3 MeV for ^{250}U, with a minute inducement by a thermal neutron, fission decay will occur. This exotic feature is due to excess of neutrons, which is not the case in the normal nuclei in the valley of stability.

Fission studies [9, 10] of ^{235}U with thermal neutrons have shown that neutron emission from the neck region is many times (order of magnitude) larger than the alpha particle emission. Since clustering probability increases at low density, alpha particle emission itself is larger compared to that of proton. Therefore, it is believed that neutron-rich neck is produced during the scission. This picture is further reinforced by the polarization effect induced by the Coulomb repulsion between the two newly formed fragment nuclei. It is also supported in microscopic studies through our calculation in relativistic mean field (RMF) theory [12] as will be shown aposteriori.
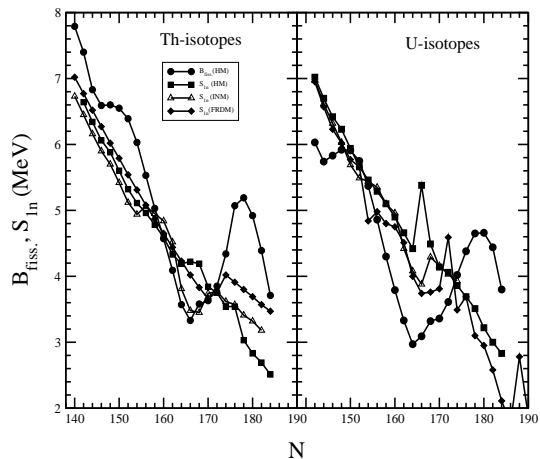

FIG. 1: Fission barrier B_f, and binding energy of the last neutron S_{1n} as a function of mass number A for Th and U isotopes. The B_f are taken from [2] and S_{1n} are taken from Refs. [2], [6], [5] and [11] for HM, INM, FRDM and RMF model respectively.
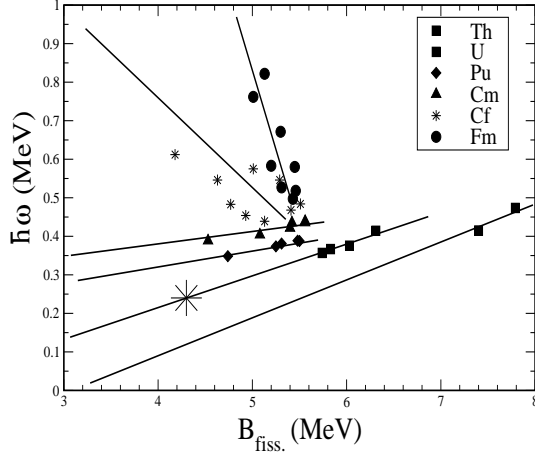
FIG. 2: The barrier curvature energy $\hbar\omega$ as function of fission barrier B_f. The B_f values and the experimental half-lives taken from Howard and Möller [2] and Ref. [8], respectively. The star on the line for Uranium in figure, denotes the value of $\hbar\omega$ to be 0.225 MeV for its $B_f = 4.3$ MeV corresponding to ^{250}U.
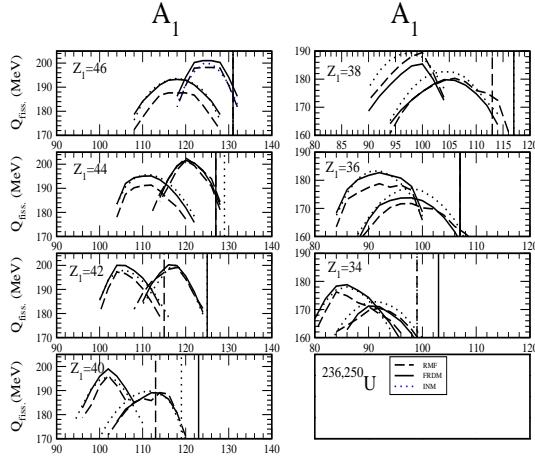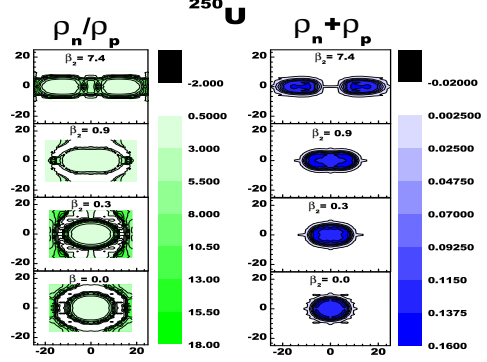

FIG. 4: The matter density distributions $\rho = \rho_n + \rho_p$ (in fm^{-3}) and $\alpha = \rho_n/\rho_p$ of ^{250}U obtained in the RMF calculations with NL3 force.

FIG. 3: $Q_{fiss.}$−value distribution for ^{236}U and ^{250}U as a function of A_1 fragment in the binary decay $A \rightarrow A_1 + A_2$. The binding energy used for calculation of Q−value is taken from [11], [4] and Refs. [6] for RMF, FRDM and INM, respectively. The vertical line marks the neutron drip-line for the corresponding element in each panel.

As is well known, the major driving force for the decay of a nucleus is the Q−value of the reaction. The probability of fragment mass yield in a given channel is directly related to the Q−value. We, therefore, calculate here the Q−value systematics of the fission of a nucleus (A,Z) decaying to (A_1, Z_1) and (A_2, Z_2) defined as $Q^f(A, Z) = BE(A_1, Z_1) + BE(A_2, Z_2) - BE(A, Z)$. In Figure 3, we have plotted the Q−values of the binary decay into two fragments A_1 and A_2, as a function of the mass number of the A_1 fragment, for all the relevant elements with even values of Z_1, starting from 34 to 46.

The complimentary fragment (Z_2, A_2) is thereby fixed. Since the yield falls rapidly with the decrease in Q−value for an element, we have only shown the distribution of Q−values lying above 90% of the highest values. For the sake of comparison, the Q−value distributions for both ^{236}U and ^{250}U are shown in Figure 3. To ensure that our conclusions remain quite general and valid, and independent of any specific mass formula, we have used the masses predicted in the three mass models RMF [11], FRDM and INM. The Q-value distributions for ^{236,250}U are presented in Figure 3 as dashed, solid and dotted curves for RMF, FRDM and INM mass formulae respectively. In the figure, three corresponding vertical lines mark the drip-lines of the respective elements predicted in the three mass models. It can be seen that the drip lines for all the three mass models agree within $\sim 3 - 5$ neutrons, except in case of Z=40 isotopic chain for the FRDM mass model. In most of the cases, the drip lines fall inside the Q−value distributions and in some cases they touch the outer fringe shown in the figure with the exception of Z=34 and 40 where the FRDM drip lines are somewhat away. Thus all the isotopes lying to the right of the drip lines will be unstable against spontaneous release of neutrons from the fragments at scission which is not the case with ^{236}U for which the drip lines are far away (See Fig. 3). In the usual fission process of ^{236}U, neutrons are emitted from the fragments after they are fully accelerated. But in the present case of ^{250}U, a certain number of neutrons will be simultaneously produced along with the two heavy fragments signaling a new mode of fission decay which may be termed as *multi-fragmentation fission*. An order of magnitude of these prompt multi-fragmentation neutrons as estimated from the mass yield plot for ^{250}U shown in Figure 3, turns out to be about 2 to 3 neutrons per fission. These are the additional neutrons apart from the normal multiplicity of neutrons emitted from the fragments. In case of

neutron-induced fission of normal ^{233}U and ^{235}U nuclei, the fission neutron multiplicities are of the order 2.5 [9]. This number is, therefore, more than doubled in case of ^{250}U fission, which will have important implications on the energetics of the fission process. This phenomenon will be more prominent in heavier Uranium isotopes due to availability of more neutrons. Although, the beta decay life-time of ^{250}U is few tens of seconds [4] it is much larger than the nucleon decay life time, which is of the order of 10^{-17} seconds and therefore it will have an implication in the $r-$process nuclear synthesis and consequently stellar evolution.

We used RMF theory with NL3 interaction to study the evolution of density as the nucleus undergoes distorsion in its shape on its path to fission, to see if microscopic calculation supports the above picture. Recently, calculations on nuclear densities obtained in the RMF studies have yielded useful insight and results on nuclear structure and dynamics [12], which we follow here. With this view we carried out RMF calculations for successively increasing deformation β_2 starting from the ground state. Now we present our results of such calculations on the total (neutrons + protons) matter density distributions $\rho = \rho_n + \rho_p$, and the ratio of neutron to proton density variable $\alpha = \rho_n/\rho_p$ as function of the deformation parameter β_2. In Figures 4, we have presented the results of such calculations for ^{250}U, with specified column on the right of the distributions showing the scale through different colours. From the figure it is clear that ^{250}U gets more and more elongated as in the usual liquid drop picture of fission and finally splits into two parts with a neck connecting them for the deformation $\beta_2 = 7.4$. It is interesting to examine the composition of the neck in this configuration. The density of the neck obtained in our calculation and also evident from the picture is $\rho = 0.02383 fm^{-3}$ which is quite low as expected. And the neutron to proton density ratio is obtained as $\rho_n/\rho_p = 3.54703$ which can be contrasted with the average density ratio of ~ 1.54 in its ground state. Thus these microscopic studies coroborate the neutron

rich-ness of neck as found in experiment and other studies [9, 10]. Similar studies for ^{236}U gives the density of the neck $\rho = 0.02440 fm^{-3}$ and the neutron-proton density ratio $\rho_n/\rho_p = 2.73089$. In case of ^{250}U, the neck is much richer in neutron since 14 extra neutrons are available compared to ^{236}U which is in conformity with expectation. This favours simultaneous emission of neutrons along with the two fragments at scission, strongly supporting the multifragmentation fission process predicted above.

In conclusion, we have identified the chain of Uranium isotopes with neutron numbers N=154 to 172 which are thermally fissile. This is a likely manifestation of the close shell nature of the magic number 162/164. We have chosen ^{249}U as a representative nucleus to study the thermal fission decay. The fission decay properties of such neutron-rich nuclei, in particular in the actinide region away from the valley of stability, have not been addressed before, which has become important in the context of RIB programs. Its fission barrier profile has been shown to be relatively flat and wide compared to ^{236}U yielding a half-life of 5.7×10^{24} years, which makes it extremely stable against spontaneous fission, while rendering it highly vulnerable to thermal neutron fission− a unique property indeed. On the basis of the probability of mass yield and microscopic RMF calculations, strong evidence of a new mode of fission decay is revealed, where in addition to two heavy fragments, 2 to 3 scission neutrons will be simultaneously emitted, which may be termed as *multifragmentation fission.* These extra neutrons are in addition to the normal multiplicity of neutrons emitted by the excited fission fragments which is of the order of 2.3 to 2.5. Thus the doubling of the neutron emission per fission will have implications for the $r-$process nucleosynthesis in the steller evolution. Whether such a nucleus presents an attractive possibility as a source of energy production needs to be examined. The above phenomenon is a general one, not restricted to ^{250}U and is likely to be more prominent in heavier Uranium isotopes.

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